

Entrained-Flow Gasification for the Utilization of Pyrolysis Oil from Mixed Plastic Waste

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Malte Hennig,* Tristan Maximilian Dreising, Anna Reeves, Teresa Oehlcke, Salar Tavakkol, Rebekka Volk, Frank Schultmann, and Dieter Stapf*



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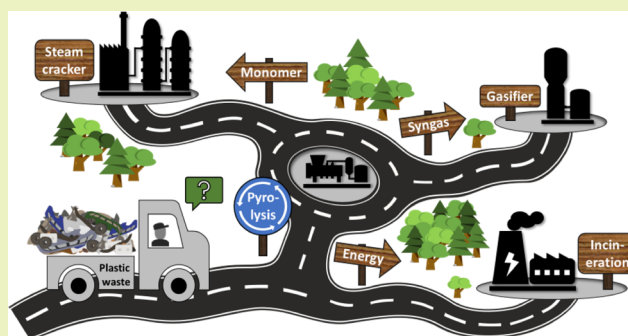
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ABSTRACT: Plastic pyrolysis aims to process plastic waste into petrochemical feedstocks by addressing waste fractions that are unsuitable for mechanical recycling due to their complex composition. Steam cracking of the resulting pyrolysis oils requires low levels of contaminants, limiting the acceptable contaminant level in feedstocks for chemical recycling. The utilization of pyrolysis oil in entrained-flow gasification (EFG) can overcome this limitation, as EFG is designed to process low-quality feedstocks such as heavy fuel oil (HFO) with high heteroatom content. For process assessment, we developed a flowsheet model for EFG of pyrolysis oil from automotive plastic waste (APW) and carried out a comprehensive life cycle assessment (LCA). HFO and APW pyrolysis oil as feedstocks for EFG show minimal differences regarding process efficiencies. In combination with the pyrolysis step, overall carbon efficiencies of 41% (methanol syngas) and 62% (Oxo syngas) are reached. Despite differing carbon efficiencies, both waste-to-syngas routes show nearly identical net climate change impacts (CCI) of 0.14 kg of CO₂e/kg of APW, indicating that the choice of syngas product composition does not significantly influence the CCI. In comparison, the established current practice of incineration with energy recovery (waste-to-energy) causes a CCI that is 1 kg of CO₂e/kg of APW higher.

KEYWORDS: plastics recycling, chemical recycling, entrained-flow gasification, pyrolysis, circular economy, life cycle assessment (LCA)



INTRODUCTION

The global production and consumption of plastics have risen steadily over the past decades and surpassed 400 million tons in 2022, contributing about 1.6 billion tons of CO₂e or 3% of worldwide greenhouse gas emissions.^{1,2} As a consequence, increasing amounts of end-of-life plastics, i.e., plastic waste, need to be managed.³ Large amounts of plastic waste remain mismanaged, leading to leakage of plastic into the environment.² Established waste management technologies, such as incineration with energy recovery (waste-to-energy), also have drawbacks, setting free additional CO₂ emissions. Meys et al. and Bachmann et al. assessed different scenarios for the plastic industry to achieve climate neutrality and operate within planetary boundaries.^{4,5} Absolute sustainability could not be achieved for the predicted quantities of plastics produced in 2030. Nonetheless, they identified the recycling of waste plastics as the crucial parameter to enable the transformation of the plastic industry toward sustainable operation.⁵ In 2020, of the 360 million tons of global plastic waste, only 9% was recycled, falling far short of the required minimum recycling rate of 75%, as calculated by Bachmann et al.^{5,6} Economic and technological restrictions of the predominant mechanical

plastic recycling pathway currently prevent higher recycling rates.^{7,8} Nevertheless, Klotz et al. determined a potential for mechanical recycling of up to 31% in a detailed material flow analysis, linking both plastic waste generation and plastic consumption in Switzerland.⁹ Chemical recycling offers another approach to increase plastic recycling rates, particularly for plastic waste that is not recyclable mechanically due to material heterogeneity or contamination.^{4,5,9–11} Pyrolysis-based chemical recycling converts plastic waste into liquid hydrocarbons as main product which can serve as feedstock for the production of monomers via conventional steam cracking (waste-to-monomers route).^{12,13} However, contaminants present in pyrolysis feedstock transfer to the hydrocarbon product. This restricts the direct use of plastic pyrolysis oil due to strict feedstock specifications regarding contaminants such

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as nitrogen, oxygen, halogens, and metals in steam cracking.^{13–16} The production of in-spec pyrolysis oil therefore requires either the restriction to cleaner feedstocks or the upgrading of pyrolysis oil to steam cracker specifications.^{16,17} By this, the advantage of processing more complex feedstocks than mechanical recycling is diminished. Another approach for chemical recycling of complex mixed plastic waste (MPW) could be the use of pyrolysis as a pretreatment step, yielding a liquid hydrocarbon feedstock suitable for utilization in entrained-flow gasification (EFG) for the production of synthesis gas. Via this route, the upgrading of pyrolysis oil is avoided altogether.¹⁸ While previous analyses have demonstrated an advantage of the waste-to-monomers route over the waste-to-energy route in terms of climate change impact (CCI), the waste-to-syngas route considered here has received little attention in the literature to date.¹⁹

In this work, we analyze the utilization of plastic pyrolysis oil derived from complex, highly heterogeneous MPW from car workshops, i.e., automotive plastic waste (APW), via the waste-to-syngas route with EFG. In order to generate the underlying mass and energy balances of the EFG process, we develop a thermodynamic flowsheet model, which we validate with data from heavy fuel oil (HFO) gasification of an industrial EFG gasifier for two scenarios: the production of methanol syngas (MeOH syngas) and the production of a syngas suitable for different oxo syntheses (Oxo syngas). We then combine the data from flowsheet modeling with the experimentally derived balances of the pyrolysis process from Stallkamp et al. to calculate the overall carbon efficiency and conduct a life cycle assessment to calculate CCIs.¹⁹ Finally, we compare the results of the waste-to-syngas routes with the waste-to-monomers and waste-to-energy routes. In a sensitivity analysis, we calculate the required minimum product yields and minimum product credits to achieve a lower CCI than the waste-to-energy route.

MATERIALS AND METHODS

The Materials and Methods section is divided into two main parts: Process Model Development and Life Cycle Assessment (LCA). These subsections describe the modeling framework, data sources, and analytical approaches used in this study. No additional experimental work was conducted beyond the experiments that have already been described and published previously.¹⁹

Process Model Development. EFG is a well-established industrial process for the production of syngas from low-quality hydrocarbon feedstocks such as HFO, bitumen, or vacuum residue, which have properties similar to plastic pyrolysis oil in terms of higher heating value and elemental composition (cf. Table 1).²⁰ In Germany, EFG is the predominant technology for the generation of MeOH syngas and Oxo syngas.^{21,22} The gasifiers are at the core of the chemical plant, producing mainly carbon monoxide (CO) and hydrogen (H₂), the building blocks for subsequent methanol synthesis or for the production of specialty chemicals such as higher alcohols, plasticizers, and detergents in Oxo syntheses.²³

In order to generate mass and energy balances for the production of MeOH and Oxo syngas from plastic waste pyrolysis oil, two flowsheet models for the EFG process are developed: (1) for MeOH syngas and (2) for Oxo syngas. The difference between the MeOH and Oxo syngas configurations arises from the different carbon monoxide (CO) and hydrogen (H₂) ratios that are required. In both cases, the H₂/CO ratio needs to be increased through the water–gas-shift reaction at the expense of CO₂ formation according to eq 1 due to the inherent hydrogen deficiency of raw syngas from EFG of HFO and similar feedstocks:^{25,26}



Table 1. Feedstock Compositions of Fuels Used in Modeling of Entrained-Flow Gasification

Feedstock	APW Pyrolysis oil	Heavy fuel oil	Bitumen
Source	19	24	20
Model purpose	Chemical recycling feedstock	Conventional syngas generation via EFG	EFG model validation
Composition (elemental, dry), wt %			
C	81.1	85.1	83.0
H	11.8	10.7	10.2
N	1.2	0.4	0.4
S	0.1	3.7	6.3
Cl	0.1	0	0
O	5.7	0	0
Ashes (incl. metals)	0	0.1	0.1 (balance)
Moisture, wt %	0.8	0	0
Higher heating value (HHV), MJ/kg	41.3	43.4	41.4

For methanol synthesis, a stoichiometric number $\text{SN} > 2$ is required,²⁵ defined as

$$\text{SN} = \frac{[\text{H}_2] - [\text{CO}_2]}{[\text{CO}] + [\text{CO}_2]} > 2 \quad (2)$$

with [H₂], [CO], and [CO₂] corresponding to the molar fraction of the respective component. A MeOH syngas free of CO₂ leads to the lowest required H₂/CO ratio of 2 that still satisfies the condition $\text{SN} > 2$ and therefore requires the lowest amount of shifting. Since CO₂ enhances reaction kinetics and increases CO conversion in the subsequent MeOH synthesis, up to 8 mol % CO₂ can be found in MeOH syngas from natural gas.^{25,26} However, due to the higher stoichiometric hydrogen demand of CO₂ in MeOH synthesis in comparison to CO and the hydrogen deficiency of HFO-based syngas in comparison to natural gas, a high CO₂ content in MeOH syngas requires additional shifting in the water–gas shift (WGS) reactor. To limit the loss of carbon through shifting, the CO₂ content in MeOH syngas is set to 1.5 mol % and $\text{SN} = 2.02$ is targeted in this study.

For Oxo syngas, an H₂/CO ratio of 1 is required.²⁶ The inert fraction in Oxo syngas should be < 0.5 mol %.²⁶ Therefore, the CO₂ content in Oxo syngas is limited to 1000 ppmv, as other inert components cannot be removed via physical scrubbing.

In gasification, two indicators are used as measures of process efficiency: Cold gas efficiency (CGE) is defined as

$$\text{CGE} = \frac{\dot{m}_{\text{Syn}} \text{LHV}_{\text{Syn}}}{\dot{m}_{\text{F}} \text{LHV}_{\text{F}}} \quad (3)$$

where \dot{m}_{Syn} and \dot{m}_{F} represent the mass flows of syngas product and feedstock, and LHV_{Syn} and LHV_{F} represent their respective lower heating values (LHVs). The CGE relates the chemical energy in the product gas to the chemical energy of the feedstock. The molar syngas yield per 100 kg of fuel represents the second indicator, defined as

$$Y_{\text{Syn}} = \frac{\dot{n}_{\text{H}_2, \text{Syn}} + \dot{n}_{\text{CO}, \text{Syn}}}{\dot{m}_{\text{F}}} \times 100 \quad (4)$$

with $\dot{n}_{\text{H}_2, \text{Syn}}$ and $\dot{n}_{\text{CO}, \text{Syn}}$ defined as the molar flow rates of H₂ and CO in the product syngas. Due to its reference to the energy content, CGE is primarily used in energy applications, e.g., integrated gasification combined cycles (IGCC), while specific syngas yield is used primarily in chemical applications where syngas composition is more important than the energy content of syngas.²⁶ Nevertheless, CGE is still used as a measure of conversion efficiency.

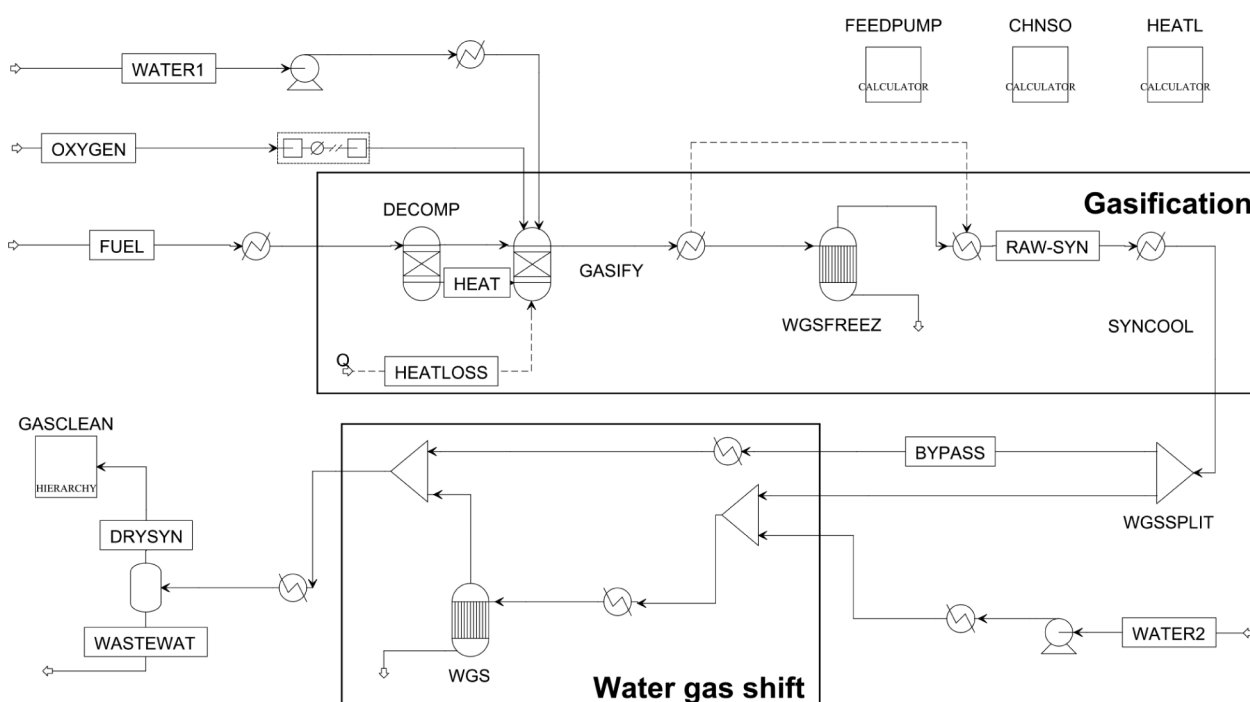


Figure 1. Aspen Plus flowsheet for the gasification of pyrolysis oil and heavy fuel oil (HFO) with a gasifier section and a water gas shift section for the adjustment of the H_2/CO ratio of raw syngas.

Gasifier Model. The process model is based on the Texaco gasifier, employing a refractory-lined gasifier in combination with a syngas cooler for recovery of high-temperature heat for steam generation.²⁶ The flowsheet model shown in Figure 1 is set up in Aspen Plus V14 (version 40.0.0.359).²⁷ The Peng–Robinson equation of state with the Boston Matthias Alpha function is used as the thermodynamic model, as this model is capable of dealing with nonpolar and mildly polar mixtures, e.g., mixtures of CO_2 , H_2S , CO , and H_2 .²⁷ The gasification process is modeled for three different fuels: pyrolysis oil from APW is part of the chemical recycling process chain, HFO is a reference feedstock for conventional syngas production, and bitumen is a feedstock for validation of the gasifier model (cf. Table 1). Pressure drop and heat loss are generally neglected unless stated otherwise. A simplified heat integration is considered based on pinch analysis to calculate minimum heating and cooling requirements, following Yang et al.²⁸ Additionally, it is assumed that any surplus heat available above 150 °C can be converted to district heat (Supporting Information). Electricity generation from excess heat via a steam cycle is not considered.

Gasification temperatures typically range between 1250 and 1450 °C to ensure close to full fuel conversion and low tar and methane content in raw syngas.^{20,26} At such high temperatures, the raw syngas composition is typically close to chemical equilibrium.^{26,29} Therefore, a chemical equilibrium approach is used for the calculation of the raw syngas composition. The oxygen-to-fuel ratio and the steam-to-fuel ratio are determined through process optimization based on the syngas yield and a minimum reactor temperature of 1350 °C (cf. Supporting Information). Following the gasification step, the raw syngas is cooled down, and the H_2/CO ratio is adjusted in a sour WGS reactor stage to meet the specifications of either MeOH or Oxo Syngas. The model allows full carbon conversion to syngas components (CO , CO_2 , CH_4), if no formation of soot is predicted based on chemical equilibrium, i.e., no inert carbon is defined ex ante. Water scrubbing for the removal of trace components (e.g., HCN , HCl , and NH_3) and the solubility of syngas components in the aqueous condensate phase are neglected. The model was validated based on industrial data from EFG of bitumen and showed close agreement between syngas composition and carbon-to-syngas conversion despite the model simplifications that were applied. For further validation, a sensitivity analysis regarding reaction temperature

and carbon conversion in EFG was carried out, underlining the model's robustness (cf. Supporting Information). A full model description is provided in the Supporting Information.

Gas Cleaning Model. The raw syngas needs to be conditioned to the respective syngas compositions set by the subsequent MeOH or Oxo synthesis; i.e., most of the CO_2 and all impurities that act as catalyst poisons need to be removed. In gasification applications, physical absorption-based processes employing methanol as a scrubbing medium—known as the Rectisol process, licensed by Linde and Lurgi/Air Liquide—are frequently used for syngas conditioning.^{30,31} Here, Rectisol scrubbing is modeled, adapted from Gatti et al.³² The process layout allows selective desorption of CO_2 (see flowsheet in the Supporting Information) if the CO_2 partial pressure in the raw syngas is sufficiently high. Thus, in the MeOH syngas case, separate recovery of CO_2 and a concentrated sour gas suitable for sulfur recovery from H_2S is possible. In the Oxo syngas case, the CO_2 desorption columns are bypassed due to the lower CO_2 loading of the raw syngas, preventing selective desorption. In contrast to the gasifier model, the PC-SAFT equation of state is used for modeling the Rectisol scrubbing.^{32,33} Activity coefficients provided by Gatti et al. are used to model the liquid-phase molecular interactions.³²

Life Cycle Assessment (LCA). LCA methodology, according to ISO 14040 and ISO 14044, is used to calculate the CCI of the modeled waste-to-syngas chemical recycling route.^{34,35} The waste-to-monomers and waste-to-energy routes have been assessed previously by the authors.¹⁹ For comparability, these assessments were updated as described in the following sections. Life cycle inventories (LCI) of all process routes are available in the Supporting Information.

Goal and Scope Definition. The goal is to identify the optimal waste treatment option in terms of climate change impact for a complex MPW that cannot be recycled mechanically. The assessment covers two chemical recycling process routes (waste-to-syngas and waste-to-monomers) as well as the established waste-to-energy technology for the treatment of MPW. Additionally, the waste-to-syngas route covers two different configurations for syngas conditioning: MeOH and Oxo syngas routes. The assessment is based on the treatment of 1 kg of MPW as functional unit. The MPW treated is APW—a mixture of different polymers (e.g., polypropylene (PP), polyethylene (PE), polycarbonate (PC), polyamides (PA)) and

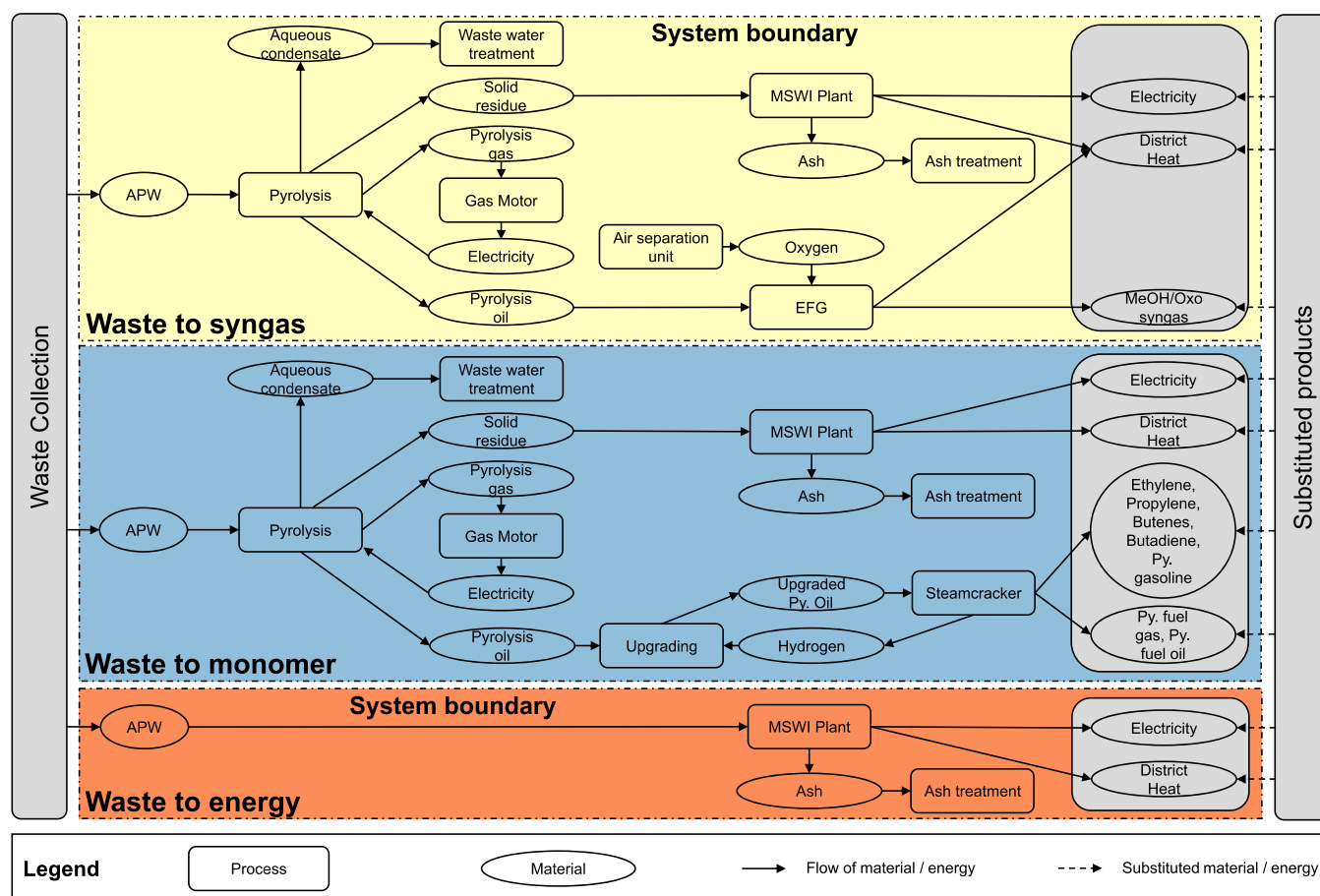


Figure 2. System boundaries of waste-to-syngas, waste-to-monomers, and waste-to-energy process routes.

nonpolymer materials (metals, glass)—for which a full characterization is provided by Stallkamp et al.¹⁹ Waste pretreatment (waste collection, recovery of metals) is excluded from the system, as previous results showed equal contribution to all waste treatment options.¹⁹ Additionally, transport of materials between processing steps is excluded due to its minimal contribution to the overall CCI and its dependence on the geographic placement and capacities of the different processing stages.^{19,36} Impacts from the construction of additional infrastructure are neglected, as either no data could be obtained or existing infrastructure is used (i.e., EFG and steam cracking plants). Modeling of all waste treatment routes assumes Germany as the geographical context.

System Boundaries. The system boundaries of the different waste treatment routes are shown in Figure 2 and are described in the following sections.

Waste-to-Syngas Process Route. MPW is pyrolyzed in a pyrolysis screw reactor. The underlying product distribution is based on experimental data.^{19,37} The utilization of pyrolysis side products is updated based on Stallkamp et al.: pyrolysis gas is utilized in a gas motor, generating electricity for heating the pyrolysis reactor and heat recovery for district heating. The solid pyrolysis residue is sent to energy recovery (see waste-to-energy process route).³⁷ Pyrolysis oil is processed by EFG to MeOH or Oxo syngas.

Waste-to-Monomers Process Route. Modeling of pyrolysis and utilization of pyrolysis side products in the waste-to-monomers process route is identical to the same steps of the waste-to-syngas process route. Following pyrolysis, the pyrolysis oil is upgraded by hydroprocessing for the removal of heteroatoms, saturation of double bonds, and aromatic compounds, and it is processed in a naphtha steam cracker for monomer production (cf. Supporting Information). Data for hydroprocessing is limited to the hydrogen demand only, due to limited process data availability.^{19,38} The modeling of steam

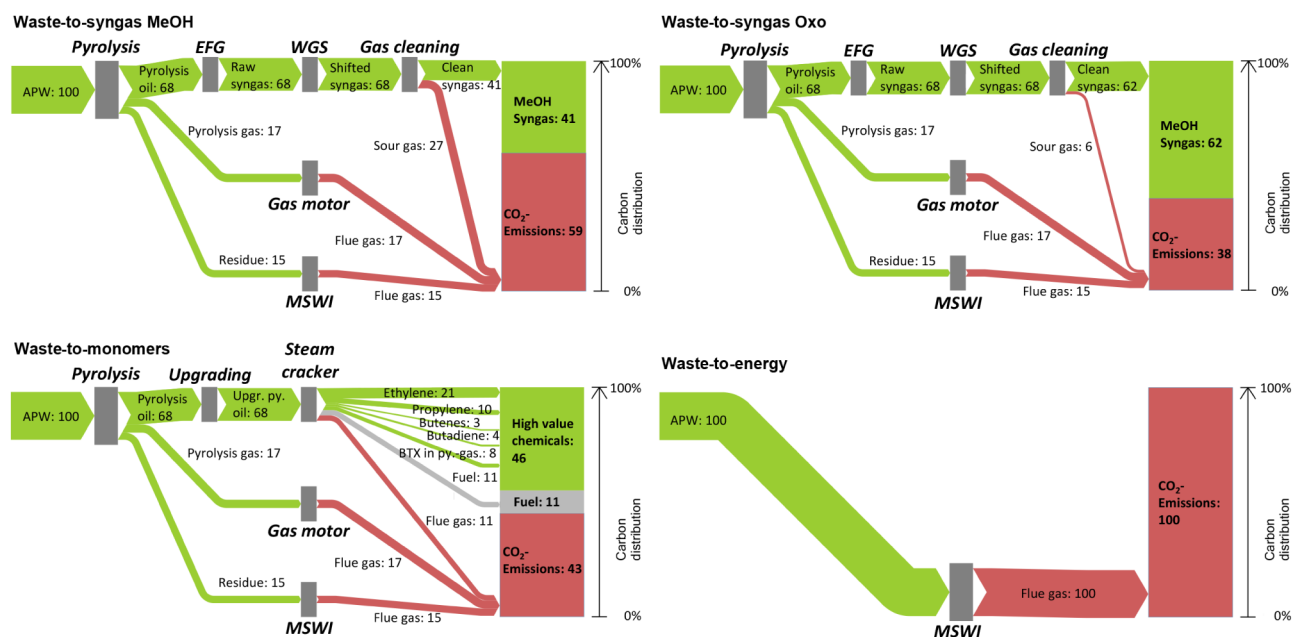
cracking from Stallkamp et al. is updated based on the data set for naphtha steam cracking from the ecoinvent v3.10 database, in accordance with the recommendations on steam cracker allocation by PlasticsEurope.^{19,39–47}

Waste-to-Energy Process Route. MPW is incinerated in a municipal solid waste incineration plant (MSWI). The model by Stallkamp et al. has been updated regarding CO₂ emissions from incineration and energy recovery efficiencies for electricity generation and district heating.^{19,48}

Impact Assessment. The impact assessment is conducted in alignment with Stallkamp et al.¹⁹ The zero-burden approach is applied, excluding any burdens from the previous life cycle for waste as a system input. Products from waste treatment are credited via the avoided burden approach (cf. Figure 2).⁴⁹ The choice of waste input as the functional unit aligns with common practice in LCA studies on waste management, enabling comparability across waste treatment technologies.^{50–52} As a reference process for the conventional production of MeOH and Oxo syngas, EFG of HFO is modeled. Credits for other products are based on the ecoinvent v3.10 database.⁵³ Details are provided in the Supporting Information. Due to limited data availability, CCI is the only impact category evaluated within the scope of this work. The CCI is calculated as global warming potential over 100 years, as defined by the sixth assessment report of the International Panel on Climate Change (IPCC)⁵⁴ via the Environmental Footprint method EF3.1.^{54,55} LCIs are modeled, and impacts are calculated with Activity Browser v2.10.3.^{56,57} Data for background activities is sourced from the ecoinvent v3.10 database.⁵³ Results from the impact assessment of the chemical recycling routes are analyzed for sensitivities regarding the yields and credits for the target products MeOH, Oxo syngas, and steam cracking products.

Table 2. Process Conditions, Process Efficiencies, and Product Yields of Entrained-Flow Gasification of APW Pyrolysis Oil for Generation of MeOH and Oxo Syngas and Conventional Syngas Generation from HFO

		MeOH syngas		Oxo syngas	
unit		APW pyrolysis oil	HFO reference	APW pyrolysis oil	HFO reference
Gasifier conditions					
Gasifier temperature	°C	1349	1350	1349	1350
Oxygen input	kg O ₂ /kg feedstock	1.02	1.00	1.02	1.00
Steam input	kg H ₂ O/kgfeedstock	0.16	0.34	0.16	0.34
Stoichiometric ratio	-	0.34	0.32	0.34	0.32
Syngas product parameters					
SN MeOH syngas	-	2.02	2.02	-	-
H ₂ /CO ratio Oxo syngas	-	-	-	1.00	1.00
LHV syngas	MJ/kg	23.2	23.4	17.4	17.5
Process efficiency parameters					
Molar syngas yield	kmol (H ₂ + CO)/100 kg fuel	12.22	12.89	12.28	12.96
Cold gas efficiency	%	81.9	80.8	84.8	83.6
Carbon efficiency ^a	%	60.4	60.2	91.7	91.4
Power demand and district heat generation					
Power demand	kWh _{el} /kg feedstock	0.79	0.80	0.72	0.73
District heating	kWh _{th} /kg feedstock	1.27	1.24	1.33	1.29

^aIncluding CO₂ for MeOH syngas configurations.**Figure 3.** Carbon flows of the waste treatment options for APW for waste-to-syngas MeOH (top left), waste-to-syngas Oxo (top right), waste-to-monomers (bottom left), and waste-to-energy (bottom right). Numbers refer to the percent of carbon input into the system. Abbreviations: MSWI—municipal solid waste incineration; Upgr. py. oil—upgraded pyrolysis oil; BTX—benzene, toluene, xylenes; py.-gas—pyrolysis gasoline.

RESULTS AND DISCUSSION

EFG of APW Pyrolysis Oil and HFO. The results from modeling APW pyrolysis oil gasification and the reference process for gasification of HFO are summarized in Table 2. A comparison between MeOH and Oxo syngas configurations shows lower carbon efficiency and lower cold gas efficiency for the MeOH configuration. The lower efficiencies result from the higher H₂ content required in the MeOH syngas, which results in additional carbon loss through CO being shifted at the expense of CO₂ formation. Since the WGS reaction is equimolar, the molar syngas yield is not affected. The comparison of pyrolysis oil and HFO as feedstocks for EFG reveals only minor differences in both the MeOH and Oxo

configurations: the higher carbon mass fraction in HFO leads to a slightly higher specific molar syngas yield. In contrast, the higher sulfur content of HFO causes a slightly lower CGE, since the heating value of H₂S is lost in sour gas that is subsequently sent to sulfur recovery (out of scope). The biggest difference between the two feedstocks becomes apparent in the significantly higher steam input required to achieve a maximum syngas yield from HFO at the selected operating temperature of 1350 °C. This effect can be traced back to the higher heating value of HFO, resulting in higher oxygen and steam input requirements for full fuel conversion. The additional steam input ultimately results in lower district heat generation. However, the choice of syngas configuration impacts the EFG performance much more than the choice of

feedstock. Therefore, the substitution of HFO with plastic pyrolysis oils of similar heating value and elementary composition in existing EFG gasifiers does not require major changes in the process layout.

In comparison to the EFG model for the production of MeOH syngas from different liquid feedstocks by Rößger et al., we calculate a slightly higher carbon efficiency for the gasification process.⁵⁸ The small difference can be ascribed to the lower gasification temperature and the model assumptions (full carbon conversion, no water prescrubbing) and provides additional validation for the model used within this work. Experimental data on the gasification of plastic waste-derived pyrolysis oil are very limited. The Carbon Cycle Lab at the Karlsruhe Institute of Technology reports successful conversion of plastic pyrolysis oil to syngas in a 5 MW pilot gasifier; however, no detailed results have been published yet.⁵⁹ Weiland et al. investigated the feasibility of combining plastic pyrolysis and EFG for a PP model plastic.⁶⁰ Due to experimental limitations (very small scale, gasification in air/nitrogen, no steam), pyrolysis oil conversion and the reported CGE are significantly lower but are expected to greatly increase for full-scale, autothermal gasifiers.⁶⁰

Carbon Flows of Waste Treatment Options. Carbon flows are calculated for all waste treatment options based on the carbon conversion efficiencies of the individual process steps, as shown in Figure 3. In combination with the carbon efficiency of the pyrolysis step (68%), the waste-to-syngas chemical recycling route reaches 41% overall carbon efficiency in the MeOH syngas configuration and 62% in the Oxo syngas configuration. The difference between both configurations is due to the adjustment of the H_2/CO ratio, which requires further shifting for the MeOH configuration. The biggest loss of carbon arises from the formation of the pyrolysis side products, pyrolysis gas and pyrolysis residue. Weiland et al. report a carbon efficiency of 48% for the gasification of PP pyrolysis oil, including the pyrolysis step.⁶⁰ This lies between the calculated carbon efficiencies for the waste-to-syngas MeOH and Oxo configurations from this work, despite the limited fuel conversion in lab-scale gasification experiments. This result highlights the need for a carbon-efficient pyrolysis pretreatment step, limiting the carbon rejection to pyrolysis side products if no other material use of pyrolysis side products is targeted or possible.^{61–63} Thus, a pyrolysis process that is optimized for a maximum pyrolysis oil yield could increase the overall carbon efficiency. Additionally, the injection of preferably green hydrogen, instead of using WGS for adjusting the H_2/CO ratio for Oxo syngas or SN for MeOH syngas, could further increase the respective carbon efficiencies of the waste-to-syngas routes.⁶⁴

The waste-to-monomer route achieves a carbon recycling rate of 46%, which lies between the two waste-to-syngas configurations. The carbon recycling rate calculated is lower compared to that of Stallkamp et al. due to the updated steam cracking model (see Supporting Information). The new model differentiates between carbon in high-value chemicals and an additional 11% of carbon that ends up in fuel applications, e.g., pyrolysis gasoline other than BTX, pyrolysis fuel gas, and pyrolysis fuel oil, in accordance with the recommendations for steam cracking allocation by PlasticsEurope.^{19,39} However, no data are available for either upgrading or steam cracking of APW pyrolysis oil, and there is little data for upgrading and steam cracking of plastic pyrolysis oils in general. Experimental data for upgrading of pyrolysis oil from waste PE showed both

a loss of product and incomplete conversion to steam cracker feedstock.⁶⁵ Additionally, steam cracking experiments with plastic pyrolysis oils that have not been upgraded previously showed altered product distributions compared to conventional naphtha steam cracking, but little information is available on the behavior of upgraded pyrolysis oil.^{15,66} As full conversion to steam cracker feedstock without loss of carbon in pyrolysis oil upgrading and no changes in steam cracking efficiency are assumed, the calculated carbon efficiency for the waste-to-monomers route should be taken as the best-case scenario and needs experimental validation.

The waste-to-energy process route converts all of the carbon from plastic waste to CO_2 and does not result in any carbon recovery.

Climate Change Impact of Waste Treatment Options.

The net CCIs, summing up all process burdens and credits, are calculated based on the LCIs presented in the Supporting Information for all waste treatment routes. Results for all waste treatment processes are listed in Figure 4. All chemical

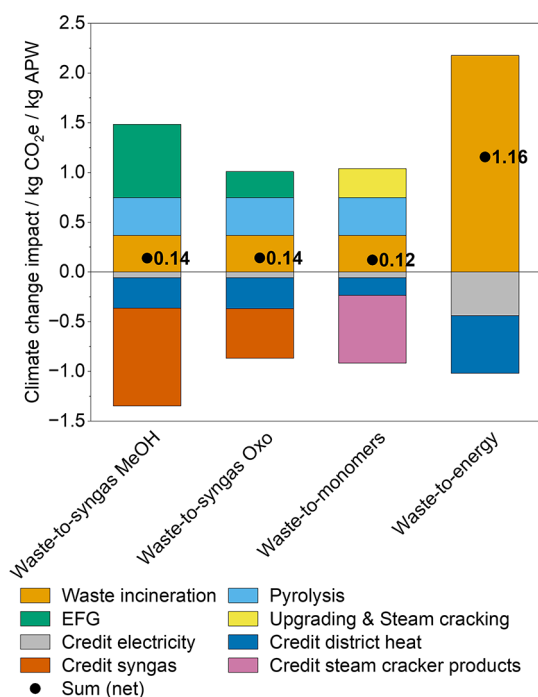


Figure 4. Climate change impacts of waste treatment options for APW for waste-to-syngas MeOH, waste-to-syngas Oxo, waste-to-monomers, and waste-to-energy.

recycling routes perform similarly, between 0.12 kg of CO_2e/kg of APW and 0.14 kg of CO_2e/kg of APW, regardless of whether steam cracking or EFG is chosen as the utilization method for pyrolysis oil. They outperform the energy recovery route by about 1 kg of CO_2e/kg of waste. The choice of syngas configuration shows no major impact, with both routes resulting in almost identical net CCIs. The higher impact of the MeOH syngas configuration in EFG is countered by equally high credits for conventional syngas generation from HFO. The pyrolysis step, including utilization of side products (categories pyrolysis and waste incineration combined), is the biggest contributor to the gross CCI (i.e., only burdens), responsible for 50% in the waste-to-syngas MeOH, 74% in the waste-to-syngas Oxo, and 72% in the waste-to-monomers routes. Upgrading and steam cracking are responsible for the

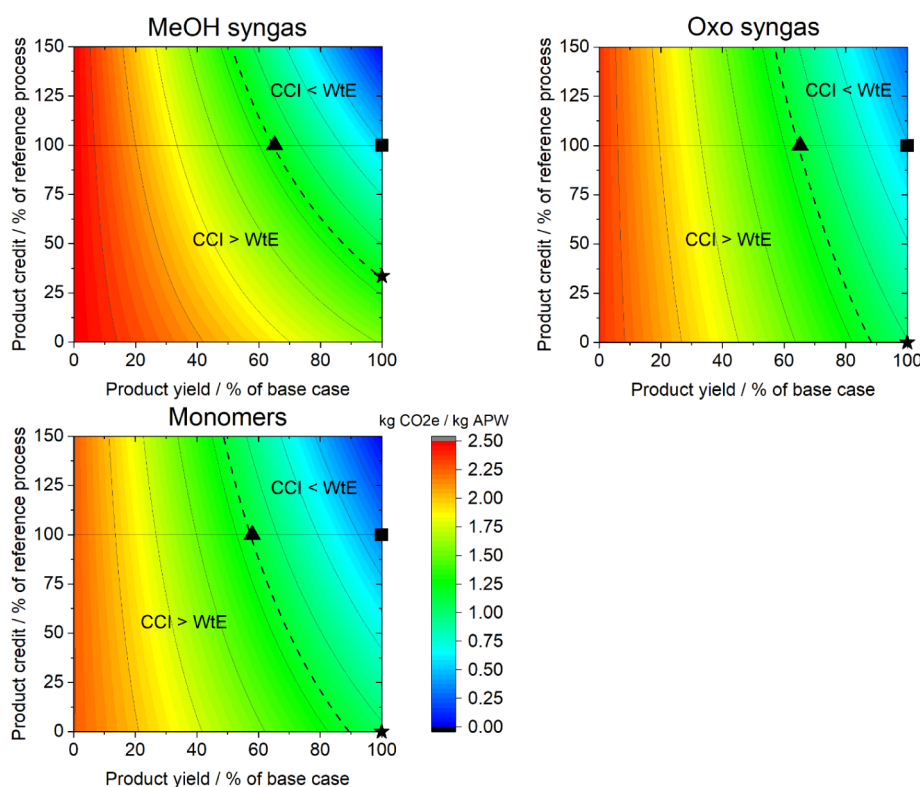


Figure 5. The effect of variation in product yield relative to the base case and product credit relative to the respective target product on the net CCI of the waste-to-syngas MeOH (left), waste-to-syngas Oxo (middle), and waste-to-monomers (right) process routes. Dashed line: Net CCI of the waste-to-energy process route. Square: Base case. Triangle: Minimum product yield. Star: Minimum credit. CCI: Climate change impact. WtE: waste-to-energy.

remaining 28% of the gross CCI of the waste-to-monomers route, splitting into 5% for upgrading and 23% for the actual steam cracking. However, due to data availability limitations, the impact of upgrading is restricted to the hydrogen demand only. The loss of carbon in pyrolysis directly translates into CO₂ emissions, resulting in pyrolysis causing the majority of the overall burdens in the chemical recycling routes. The main sources for credits are MeOH syngas (73% in waste-to-syngas MeOH) and Oxo syngas (57% in waste-to-syngas Oxo). Steam cracker products are responsible for 74% of the credits of the waste-to-monomers route, of which approximately 95% result from the substitution of steam cracker main products and 5% from the substitution of fuels. District heat is responsible for 23% of the credits (waste-to-syngas MeOH), 36% (waste-to-syngas Oxo), and 20% (waste-to-monomers). Electricity makes up 4% to 7% of the overall credits of the chemical recycling routes. In comparison, electricity is responsible for 43% and heat is responsible for 57% of credits in the waste-to-energy route. Generally, the preference for chemical recycling over energy recovery when producing chemical feedstocks is in line with the literature, as recently outlined in a review by Zhang and Nakatani.⁶⁷ For the combination of pyrolysis with EFG of plastic pyrolysis oil, no comparable LCA is known to the authors due to differences in materials, technologies, and methodological choices, such as system boundaries. However, Arena et al. and Voss et al. concluded that the direct gasification of MPW and municipal solid waste is preferable to incineration.^{50,68} In a larger context, Keller et al. showed that the combination of pyrolysis and EFG for the production of methanol leads to a reduction in CCI, aligning with the results of this study.²¹ The waste-to-syngas route for heterogeneous,

complex MPW therefore shows a significant CCI reduction potential. For example, 1.4 million tons of sorting residues from lightweight packaging waste were incinerated in Germany in 2020.⁶⁹

Sensitivity Analysis. The advantage of chemical recycling routes over the waste-to-energy route results from two effects: (1) carbon is retained in a product rather than emitted in the form of CO₂ and (2) these products are credited with the respective avoided burdens.⁵⁰ Together, these effects contribute to a reduced CCI. In this waste-based (input-based) LCA, these effects correspond to (1) the achievable product yield of carbon retained in MeOH syngas, Oxo syngas, and monomers, and (2) the value of the credit assigned to these target products. To determine the required minimum product yield and minimum credit to achieve a lower net CCI than the reference case waste-to-energy, detailed process models are required. These models must correlate the product yield with CCI across multiple operating conditions. However, such models are currently unavailable for key processes, such as pyrolysis, upgrading, and steam cracking of the pyrolysis oil. In consequence, the sensitivity analysis in this study uses a static, model-free approach. This approach utilizes aggregated data across the entire process chain, based on the results obtained from the respective base cases (cf. Figure 3 and Supporting Information). The model-free approach takes into account the gross CCI of the respective chemical recycling route, i.e., considering only burdens, along with the product yields and credits of the target products MeOH syngas, Oxo syngas, and monomers. Credits from electricity or district heating are excluded due to their significant reduction in recent years as a result of defossilization efforts in the German energy

system.^{70,71} This ensures a conservative assessment. A product yield of less than 100% indicates carbon losses along the chemical recycling process chain relative to the base case, which are treated as emissions of the respective amount of CO₂. Yields above 100% are excluded, as such values would be speculative within the limitations of the static model used. Further details regarding the calculation of the sensitivity analysis are provided in the [Supporting Information](#).

The results show that, for the waste-to-syngas Oxo route and the waste-to-monomers route, the effect of carbon retention alone is sufficient to achieve a lower CCI than the waste-to-energy route ([Figure S](#), star symbols). While product credits further improve the performance of these routes, they are not essential to surpass the energy recovery benchmark. In contrast, for the waste-to-syngas MeOH route, a minimum credit of 0.49 kg of CO₂e/kg of MeOH syngas is required at 100% product yield (base case) to match the CCI of the waste-to-energy process route.

Additionally, the minimum product yield required to achieve equivalence with the energy recovery route—based on the reference process credits—can be determined (cf. [Figure S](#), triangle symbols): for waste-to-syngas MeOH and Oxo routes, at least 65% of the base case product yield has to be reached. In the EFG process model sensitivity analysis (cf. [Supporting Information](#)), the maximum yield loss due to variations in reactor temperature and carbon conversion was found to be 4%. Thus, even with such deviations between the process model and an industrial implementation, performance would likely remain superior to that of energy recovery.

If the required minimum product yield of the waste-to-monomers route of 58% - the lowest among the chemical recycling routes—can be reached, depends on the actual yields of pyrolysis, upgrading of pyrolysis oil, and the subsequent steam cracking. While yield prediction of these processes remains challenging due to their dependence on complex reaction kinetics, the pyrolysis oil yield used in this study is based on pilot-scale experimental data specific to the employed MPW feedstock.^{12,72} However, data for the upgrading and steam cracking of plastic pyrolysis oils, particularly those derived from complex MPW, remain limited.^{15,19,38} Early-stage experimental results showed significant variability in steam cracking yields from untreated pyrolysis oils, highlighting the need for further validation using experimental data.^{15,73} Consequently, the required minimum product yield implies that upgrading and steam cracking of the plastic pyrolysis oil must achieve approximately 60% of the product yield of fossil naphtha steam cracking to ensure the waste-to-monomers route performs better than the waste-to-energy route.

Among all of the modeled process steps, pyrolysis accounts for the highest loss of carbon, making it the most critical target for optimization. Such optimization requires further experiments to determine the change in the overall product distribution and product composition, taking into account both the overall product yield as well as pyrolysis oil quality, i.e., HHV and contaminant content.^{63,74,75} Both factors affect the efficiencies of downstream EFG, upgrading, and steam cracking processes, thereby determining the overall product yield.

The CCI of the energy recovery route is sensitive to future reductions in the CCIs of electricity and district heat generation. As Germany continues to decarbonize its energy system in line with national emission reduction goals, the CCI of energy recovery is likely to increase gradually in the

future.^{70,71,76} Consequently, in accordance with Voss et al., the advantage of the chemical recycling routes will grow over time.⁵⁰ As a result, the required minimum product yields and credits will decrease, expanding the operational feasibility of chemical recycling technologies.

Study Limitations. The study results underlie limitations and uncertainties due to (1) model assumptions, availability, and accuracy of data, and (2) the applied LCA methodology. For compiling the LCIs, data from pilot-scale experiments, thermodynamic modeling, and literature were used. The validity of the data is limited to the waste used as feedstock within this case study. However, similar behavior is expected for other mixed plastic wastes with a comparable elemental composition. To account for model simplifications in the EFG process model, key parameters were evaluated for their impact on process efficiency within a sensitivity analysis. Due to a lack of data for upgrading and cracking of pyrolysis oil from mixed plastic waste, proxy processes, approximations, and simplifications were necessary. No optimization for maximum performance in terms of CCI was carried out, either for the entire process chain or for individual process steps of the chemical recycling routes, due to the extensive additional experimental work that would be required. Thus, LCIs should be validated on an industrial scale once the respective processes are established.

The LCA is limited to the waste perspective, excluding any product-based analysis, due to the focus on the comparative assessment of different waste treatment options. A product-based analysis of the different waste treatment routes would require the definition of a combined functional unit that takes into account both the different target products as well as the treatment of mixed plastic waste.⁵¹ However, a transformation to a product-based approach would allow the inclusion of our model into product-centered system-level models, such as presented by Meys et al. and Bachmann et al.^{4,5}

CONCLUSION

In this work, we analyzed the potential of entrained-flow gasification (EFG) for the utilization of pyrolysis oil from complex mixed plastic waste from car workshops. A flowsheet model for MeOH and Oxo syngas production was used to calculate the mass and energy balances. The similar behavior of APW pyrolysis oil and conventional HFO, the standard feedstock for syngas production via EFG in Germany, is evident. For the EFG of APW pyrolysis oil, carbon efficiencies of 60% for MeOH syngas and 91% for Oxo syngas were calculated. In combination with the pyrolysis conversion step, a total carbon efficiency of the waste-to-syngas process route of 41% (MeOH syngas) and 62% (Oxo syngas) is achieved. The carbon efficiency of the waste-to-monomers process route (46% carbon material use, 11% in fuels) lies between the two waste-to-syngas routes. Energy recovery does not result in any carbon recovery (0%). Thus, by comparing all process routes, the production of Oxo syngas achieves the highest carbon efficiency. When considering the CCI of the process routes, all chemical recycling routes outperform the waste-to-energy route by 1 kg of CO₂e/kg of APW at a CCI of 0.14 kg of CO₂e/kg of APW. While MeOH syngas production results in higher carbon loss, this loss is offset by credits from conventional MeOH syngas production, meaning syngas configuration does not affect the CCI.

The highest potential for improving carbon efficiencies, and thus lowering CCI, lies in the optimization of the pyrolysis

process depending on the subsequent utilization of the resulting pyrolysis oil. For the waste-to-monomers route, an optimization of the pyrolysis step toward higher-quality pyrolysis oil may be beneficial. In contrast, for the waste-to-syngas route, the pyrolysis process could be tuned toward higher pyrolysis oil yields, as EFG is not impacted significantly by contaminants. Material use of the remaining pyrolysis side products would further enhance carbon efficiency.

The sensitivity analysis indicates a sufficiently broad parameter space of minimum product yields and minimum product credits in which the chemical recycling routes outperform the established waste-to-energy route in terms of CCI. However, experimental validation is essential to reduce uncertainties related to EFG, pyrolysis, and particularly the upgrading and steam cracking of pyrolysis oils to ensure accurate LCIs. Future research should focus on the economic implications of the combination of EFG and pyrolysis. Finally, the impact of waste quality on the different pyrolysis oil utilization pathways in terms of process efficiencies and its correlation with waste characteristics, e.g., heating value and heteroatom content, needs to be analyzed.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acssuschemeng.5c03932>.

Additional information on the process model, sensitivity analysis, and life cycle inventories (PDF)

■ AUTHOR INFORMATION

Corresponding Authors

Malte Hennig – Institute for Technical Chemistry (ITC), Karlsruhe Institute of Technology (KIT), Karlsruhe, Baden-Württemberg 76131, Germany; orcid.org/0000-0002-0289-5608; Phone: +49 721 608 24351; Email: malte.hennig@kit.edu

Dieter Stapf – Institute for Technical Chemistry (ITC), Karlsruhe Institute of Technology (KIT), Karlsruhe, Baden-Württemberg 76131, Germany; orcid.org/0000-0001-6499-062X; Phone: +49 721 608 29270; Email: dieter.stapf@kit.edu

Authors

Tristan Maximilian Dreising – Institute for Technical Chemistry (ITC), Karlsruhe Institute of Technology (KIT), Karlsruhe, Baden-Württemberg 76131, Germany; orcid.org/0009-0007-0465-449X

Anna Reeves – Institute for Technical Chemistry (ITC), Karlsruhe Institute of Technology (KIT), Karlsruhe, Baden-Württemberg 76131, Germany

Teresa Oehlcke – Institute for Industrial Production (IIP), Karlsruhe Institute of Technology (KIT), Karlsruhe, Baden-Württemberg 76131, Germany; orcid.org/0009-0001-0037-9415

Salar Tavakkol – Institute for Technical Chemistry (ITC), Karlsruhe Institute of Technology (KIT), Karlsruhe, Baden-Württemberg 76131, Germany; orcid.org/0000-0001-5142-2439

Rebekka Volk – Institute for Industrial Production (IIP), Karlsruhe Institute of Technology (KIT), Karlsruhe, Baden-Württemberg 76131, Germany; Department of Sustainable Systems Engineering (INATECH), University of Freiburg

Freiburg, Baden-Württemberg 79110, Germany;

orcid.org/0000-0001-9930-5354

Frank Schultmann – Institute for Industrial Production (IIP), Karlsruhe Institute of Technology (KIT), Karlsruhe, Baden-Württemberg 76131, Germany; orcid.org/0000-0001-6405-9763

Complete contact information is available at:

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Notes

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■ ABBREVIATIONS

APW, automotive plastic waste; BTX, benzene, toluene, xylenes; CCI, climate change impact; CGE, cold gas efficiency; EFG, entrained-flow gasification; HFO, heavy fuel oil; HHV, higher heating value; IGCC, integrated gasification combined cycle; LCA, life cycle assessment; LCI, life cycle inventory; LHV, lower heating value; MeOH, methanol; MPW, mixed plastic waste; MSWI, municipal solid waste incineration; PA, polyamide; PC, polycarbonate; PE, polyethylene; PP, polypropylene; py, pyrolysis; SN, stoichiometric number; Upgr, upgraded; WGS, water–gas-shift reaction; WtE, waste-to-energy

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